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Development of NDA Methods for Neptunium Metal

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ABSTRACT

Many techniques have been developed and applied in the US and other countries for the control of the special nuclear materials (SNM) plutonium and uranium, but no standard methods exist for the determination of neptunium in bulk containers. Such methods are needed because the U.S. Department of Energy requires all Government-owned ^{237}Np be treated as if it were SNM and the International Atomic Energy Agency is considering how to monitor this material. We present the results of the measurements of several samples of neptunium metal with a variety of techniques. Analysis of passive gamma-ray spectra uniquely identifies the material, provides isotopic ratios for contaminants, such as ^{243}Am, and may provide information about the shielding, mass, and time since processing. Active neutron interrogation, using the delayed neutron technique in a package monitor, provides useful data even if the neptunium is shielded. The tomographic gamma scanner yields a map of the distribution of the neptunium and shielding in a container. Active photon interrogation with pulses from a 10-MeV linac produces delayed neutrons between pulses, even when the container is heavily shielded. Data from one or more of these techniques can be used to identify the material and estimate a mass in a bulk container.

INTRODUCTION

Many techniques have been developed and applied in the US and other countries for the control of the special nuclear materials (SNM) plutonium and uranium, but no standard methods exist for the determination of neptunium in bulk containers. Such methods are needed because the U.S. Department of Energy requires all Government-owned ^{237}Np be treated as if it were SNM and the International Atomic Energy Agency is considering how to monitor this material. Virtually all spent fuel and reprocessing waste contain ^{237}Np, and the inventories continue to grow. Historically, ^{237}Np was separated in only small quantities as target materials for ^{238}Pu production and for smoke detectors, neutron generators, and research. Recent research efforts have focused on removing these long-lived actinides from highly radioactive waste to facilitate disposal. The resulting separated ^{237}Np could encourage commercial applications and international commerce in this material. Neptunium 237, as well as americium, has been the focus of recent proliferation concerns [1].

In this report we describe results to date of the application to ^{237}Np metal of four Non-Destructive Assay (NDA) techniques that were originally developed for plutonium and uranium: passive gamma-ray spectroscopy, the package monitor, the tomographic gamma scanner, and active interrogation with a linac.

PASSIVE GAMMA-RAY SPECTROSCOPY

The gamma-ray lines at 300, 312, 341, 376, 399, and 416 keV from the daughter ^{233}Pa ($T_{1/2}$ = 27.0 days) are used for the analysis of neptunium samples. Neptunium samples are often stored with lead shielding in the container because of the potential radiation exposure of personnel produced by these lines. The lines directly from neptunium, such as the 143 and 151 keV lines, are weak and too

low in energy to penetrate the lead. Use of the strong ^{233}Pa lines assumes that the time since processing is at least three half lives (81 days) so that equilibrium has been achieved. Lines from other isotopes, such as ^{243}Am, are used to calculate the isotopic composition of the sample.

Figure 1 shows a typical spectrum from a 3-g sample of ^{237}Np metal. The measurement was made with a 26.4%-efficient germanium detector that had a resolution of 1.81 keV at 1332 keV. The sample was placed 2 m from the detector to reduce the dead time. No filter was used because some useful gamma-ray lines were at low energy.

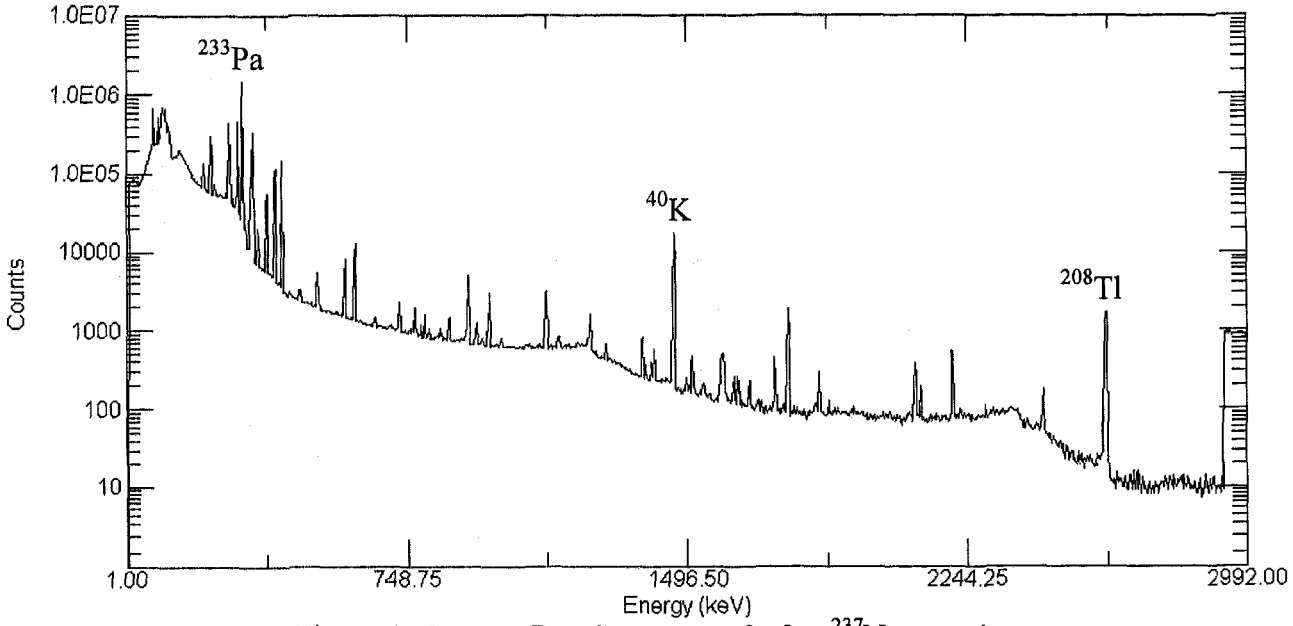

Figure 1. Gamma-Ray Spectrum of a 3-g ^{237}Np sample

The analysis program GammaVision from EG&G ORTEC was used to locate more than 300 gamma-ray lines in this spectrum. The strong ^{233}Pa lines are at 300, 312, 341, 376, 399, and 416 keV. The isotope ^{239}Np ($T_{1/2}$ = 2.355 days) from the decay of ^{243}Am in the sample produces lines at 228 and 278 keV. The 74.7-keV line from ^{243}Am is often not useful because any lead shielding present attenuates this low energy line and produces interference from the Pb $K_{\alpha 1}$ x-ray at 75.0 keV. The isotope ^{241}Am produces a line at 59.5 keV. Most of the higher energy lines are from natural background.

The analysis program FRAM [2] was used to calculate the isotopic composition. The program was originally written to determine the isotopic composition of plutonium samples to high precision, and neptunium was treated as an impurity (<10%). We have modified the parameters in the program to analyze ^{237}Np metal. For the data shown in Figure 1 the program calculates that the mass ratio ^{243}Am/^{237}Np is $0.1549 \times 10^{-3} \pm 0.13\%$ and indicates that the masses of other isotopes are negligible. We do not have independent confirmation of the assay for this sample or any other sample at present because the available samples are old and poorly documented. We are awaiting shipment of better characterized samples.

PACKAGE MONITOR

The package monitor is an active neutron interrogation system designed to rapidly search containers, packages, and luggage for SNM [3,4]. The neutrons are produced by a pulsed neutron generator using the D(t,n)^{4}He reaction. The initial neutron energy of 14 MeV is rapidly (< 200μs) reduced to thermal energy by scattering in the graphite and polyethylene lined walls of the package monitor chamber. Arrays of cadmium covered ^{3}He detectors embedded in the walls of the chamber detect fission neutrons after the pulse. The chamber can accommodate packages as large as 1 × 1 × 1 m^3, and the absolute neutron detection efficiency is approximately 6%. Interrogation times for detection are less than one minute for HEU.

Figure 2 shows the number of neutrons detected as a function of the time after the interrogating pulse. The ^{237}Np curve is lower than those for ^{239}Pu and ^{235}U but higher than the curve for an empty chamber. The ^{237}Np curve is very similar to the one for ^{238}U. The signal for ^{237}Np in the region of interest (ROI) 3 to 9 msec after the pulse is 56 sigma above the signal for the empty chamber. Additional information from another technique, such as passive gamma spectroscopy, can distinguish ^{237}Np from ^{238}U.

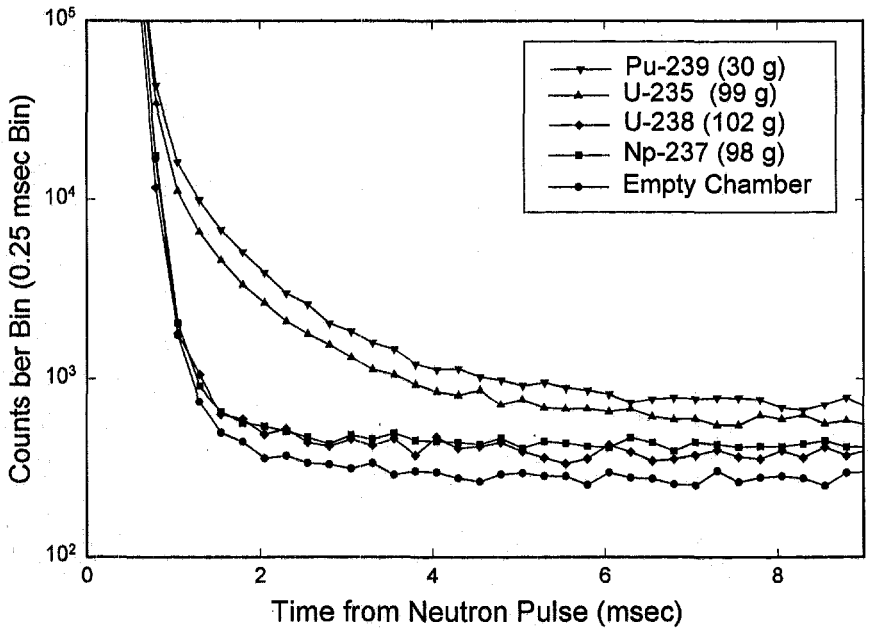

Figure 2. Data from the active interrogation package monitor.

The curves in Figure 2 demonstrate two different data analysis techniques provided by the package monitor. The ^{239}Pu and ^{235}U curves demonstrate the differential die-away technique, in which thermalized neutrons do the interrogation for the materials. The thermalized neutrons die away much more slowly than the fast and epithermal neutrons, hence the name differential die-away technique. The slow decay is evident between 1 and 3 msec. The ^{237}Np and ^{238}U curves demonstrate the delayed neutron technique. The fast and epithermal neutrons provide the interrogation. Thermal neutrons cannot cause ^{237}Np and ^{238}U to fission because they have thresholds at 600 keV and 1 MeV, respectively. The neutrons seen after the pulse are delayed neutrons.

Shielded ^{237}Np can also be measured because the 14 MeV neutrons from the pulsed neutron generator are very penetrating. For example, the signal in the 3 to 9 msec ROI for a bare 98-g ^{237}Np sample was 22 sigma above background and 33 sigma when shielded by 5.08 cm lead. The increase is attributed to (n,2n) reactions in lead, thus producing more interrogating fast and epithermal neutrons close to the sample. The signal with just lead present was equal to background. Measurements were also made with neutron shielding, such as polyethylene.

TOMOGRAPHIC GAMMA SCANNER

We have applied a tomographic gamma scanner, which was originally developed to assay SNM in 208-liter drums [5], to image samples of ^{237}Np in various configurations. The strongest emission gamma rays are from ^{233}Pa at 300, 312, 341, 376, 399, and 416 keV. Two measurements of each configuration were used to construct images: 1) a transmission measurement with ^{137}Cs and ^{133}Ba transmission sources and the ^{237}Np sample(s) present, and 2) an emission measurement with the ^{237}Np sample(s) present but without the ^{137}Cs and ^{133}Ba sources. The transmission measurement provides information about gamma-ray attenuation by any matrix material as well as the ^{237}Np source itself. Measurements were made with the ^{237}Np samples bare (except for thin-walled containers to control contamination) or shielded. As many as four ^{237}Np samples were positioned at various radial and vertical distances using vertical aluminum tubes in 208-liter drums, either empty or containing various matrices.

Figure 3 shows four small (less than 1 g each) ^{237}Np samples in an empty 208-liter drum. The transmission measurement and the emission measurement each required 1600 sec, for a total of 3200 sec. The drum was scanned in sixteen layers with a 3.175 cm (diameter) × 16.2 cm (length) collimator. A lead filter 0.95 cm thick was placed in front of the germanium detector to reduce the dead time to less than 20%. All four sources are visible. One of the sources (the lowest in the drum) is approximately four times stronger than the other three, and therefore appears larger at the intensity scale used for the display.

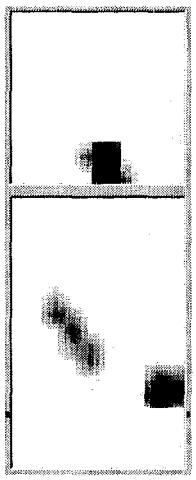

Figure 3. TGS emission image of four ^{237}Np sources at different heights and radial positions with a 208-liter drum. The upper image is a tomographic slice view (horizontal slice) through the strongest source and rotated to center the source; the lower image is a "radiograph" side view.

Scans of other drums provided information on the effects of different matrices and shielding. The small ^{237}Np samples were still visible when the drum was filled with small polyethylene beads. Investigations of higher Z matrices are planned. Nested cylindrical lead shields were used to investigate the sensitivity as a function of shielding thickness closely surrounding each ^{237}Np source.

ACTIVE INTERROGATION WITH A LINAC

This method uses an external radiation probe of 10-MeV bremsstrahlung photons to induce fission of the ^{237}Np sample and thus produce delayed neutrons. We originally developed this delayed neutron method for the investigation of highly enriched uranium (HEU) systems [6]. A brief report on the application to ^{237}Np emphasizing the physics of multiplication has been presented elsewhere [7]. The photon source, a 10 MeV electron linear accelerator, is operated in a pulsed mode with a pulse width of ~6 microseconds at a frequency of ~50 Hz. For all the measurements 45,000 pulses from the linac were used. The linac output was approximately 150 R/min at one meter from the bremsstrahlung source. Neutrons are detected by an array of ^{3}He tubes embedded in polyethylene slabs above and on two sides of the material being interrogated. The data acquisition is gated off during the linac beam burst and for an additional 2000 microseconds. The neutron detection times are recorded, and subsequently analyzed with the Feynman reduced variance method [8,9,10]. This analysis provides a measure of the "singles" rate (N1/sec) and "doubles" rate (N2/sec) for neutron events detected from fission events. These fission events are predominantly produced by the delayed neutrons from fission products resulting from the interaction with the 10 MeV bremsstrahlung photons during the interrogating probe burst.

Eleven measurements were made on a 98-g ^{237}Np metal sample. For comparison, 11 measurements were made on a 100-g ^{235}U (93% HEU) sample and 5 on a 1004-g ^{238}U (depleted uranium) sample. Nine were made with a ^{252}Cf neutron source to determine the neutron detection probability for fission energy neutrons and to determine the effect of the linac photon burst on the performance of the detector. The different measurements involved different shielding configurations of polyethylene, lead, and iron.

The results of one measurement for ^{237}Np are shown in Figure 4. The N1/sec values (diamonds) show a sharp decrease from the earliest times to about 4000 microseconds after the beam burst

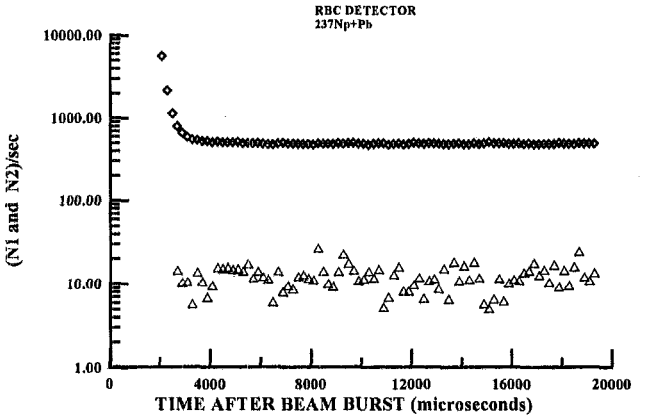

Figure 4. The N1/sec values (diamonds) and the N2/sec values (triangles) as a function of time after the linac beam burst for a 98-g ^{237}Np sample.

whereas the N2/sec values (triangles) remain essentially constant. The N2/sec values are constant because, as noted above, they are fission neutrons produced by delayed neutrons, which are essentially constant. The ^{237}Np sample was contained within a lead cylinder with 1.6 cm thick walls to mitigate any possible effect of the large gamma-ray background from the ^{233}Pa daughter nuclei on the neutron detector.

Table 1 shows the neutron counting rates in the time interval 8000 to 16000 microseconds after the burst for some selected configurations. The rates decrease as the thickness of polyethylene (CH_2) increases because the emitted neutrons are absorbed. Lead and iron decrease the rate because the incident photons are attenuated. As noted above, ^{237}Np is often stored with lead shielding because of the radiation from the daughter ^{233}Pa. Placing a block of beryllium ($7 \times 7.5 \times 10$ cm^3) in the photon beam at the exit window of the linac, with the 10-cm dimension along the beam, to convert the photon beam to a neutron beam resulted in a lower rate. However, surrounding the sample with a beryllium shell more than doubled the rate. The rates for ^{235}U and ^{238}U, discussed above, are also given.

Table 1. Neutron Counting Rates

Material	N1/sec	N2/sec
^{237}Np(98 g)+Pb(1.6 cm)	496.9 ± 1.14	12.0 ± 0.8
^{237}Np(98 g)+Pb(1.6 cm)+CH_2(6.35 cm)	208.5 ± 0.8	7.5 ± 1.0
^{237}Np(98 g)+Pb(1.6 cm)+CH_2(8.89 cm)	75.6 ± 0.4	3.2 ± 0.2
^{237}Np(98 g)+Pb(1.6 cm)+Fe(6.35 cm)	248.3 ± 0.7	9.6 ± 0.6
^{237}Np(98 g)+Pb(1.6 cm)+Fe(6.35 cm)+Be converter block ($7 \times 7.5 \times 10$ cm^3)	178.9 ± 0.9	7.0 ± 0.4
^{237}Np(98 g)+Pb(1.6 cm)+Be spherical shell(10.16 cm ID, 17.78 cm OD)	1110 ± 3	35.0 ± 1.2
^{235}U(100 g)+Cd+Pb(1.6 cm)	407.9 ± 1.0	17.7 ± 0.5
^{238}U(1004 g)+Pb(1.6 cm)	2996 ± 5	10 ± 3

The results for the comparison measurements on ^{235}U and ^{238}U are also included in Table 1. The ^{235}U and ^{238}U samples were placed within the same lead shield as the ^{237}Np sample. A cadmium cylinder also surrounded the ^{235}U sample to prevent fission events induced by low energy neutrons. For ^{235}U the N1/sec and N2/sec values are similar to those for ^{237}Np. For ^{238}U the N1/sec value is approximately an order of magnitude larger than those for ^{237}Np (except when surrounded by a beryllium spherical shell) and ^{235}U, due to the larger mass of the ^{238}U sample. The N2/sec value for the ^{238}U sample is small when normalized to mass, reflecting the poor overlap between the delayed neutron energy spectrum and the neutron induced fission cross-section for ^{238}U. For both the ^{237}Np and ^{235}U samples significant values of N2/sec are observed, indicative of neutron multiplication, that is, the delayed neutrons from fissions product initiate fission events. Thus, we are easily able to discriminate between ^{238}U and ^{237}Np or ^{235}U. However, another technique, such as measurement of a passive gamma-ray spectrum is required to discriminate between ^{237}Np and ^{235}U.

DISCUSSION

Major goals of NDA include the identification of a sample and the determination of its mass and composition. Passive gamma-ray spectroscopy provides identification and composition, if the sample is not heavily shielded. TGS provides information about the shape and distribution of the radioactive material and the shielding. TGS also provides a gamma-ray spectrum because it contains a germanium detector. The package monitor and the linac technique, which are based on detecting neutrons, can easily detect shielded SNM. For the most heavily shielded samples, the linac is most suitable of the four techniques because the high energy photon beam is very penetrating. All four techniques provide a response that increases with mass. However, the response also depends on shielding, self-attenuation, geometry, and chemical composition. The most accurate strategy is the use of calibration samples with the same shielding, geometry, and chemical composition. If this is not possible, information from two or more techniques plus other known information can be combined to determine the mass. Some calculations, such as correcting for shielding, may be required.

The choice of techniques often depends on the size and complexity of the required equipment. Passive gamma-ray spectroscopy can be performed with a portable germanium detector system. Usually the sample must be moved to a low background area. The package monitor and the TGS are not portable so that samples must be brought to them. The linac hardware is the largest and most complex and requires a special facility because of the high radiation produced. All of the measurement techniques require less than one hour, the TGS being the slowest.

We have focused on NDA of neptunium metal to meet known user requirements. Our techniques are also applicable to neptunium oxide and waste. We are soliciting input from users on their requirements.

CONCLUSIONS

We have described results to date from four techniques for NDA of ^{237}Np metal. No one technique is usually sufficient for a complete assay without additional information. Passive gamma-ray spectroscopy usually provides the most detailed information about the composition. TGS provides the spatial distribution and an estimate of the mass. The package monitor and linac technique provide information when heavy shielding is present. For all techniques best accuracy is achieved with calibration standards closely matched to the samples to be assayed.

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